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East Europe Report

SCIENTIFIC AFFAIRS

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ARTICLE EXAMINES PRESENCE OF KR-85 IN ATMOSPHERE

Budapest FIZIKAI SZEMLE in Hungarian No 2, Feb 1969 pp 33-35

[Article by Eva Csongor, Nuclear Research Institute of the Hungarian Academy of Sciences, Debrecen: "Kr-85 in the Atmosphere"]

[Text] Atmospheric Radioactivity

The radioactive material content of the atmosphere is partly of natural origin and partly the result of human activity.

The natural radioactivity in the air can be attributed essentially to the decay products of U-238, Th-232 and U-235 occurring in the earth's crust, which get into the atmosphere by means of diffusion (Rn, Tn and their decay products). Also of natural origin is that smaller part of atmospheric activity which comes into being as a result of the mutual effect of cosmic radiation and the atoms of the atmosphere (H-3, Be-7, C-14, Na-22, etc). But the radioactivity of the atmosphere makes up only about 1 percent (1 mrem per year) of that radiation load a person receives as a result of soil radiation and cosmic radiation. The radiation of the environment represents a dose on the order of 100-150 mrem per year for the human organism. These data, however, depend greatly on geological factors; for example, the dose could be 10 times this value near Uranium deposits.

The radioactivity of the atmosphere has increased gradually in the past 15 years as a result of human activity as artificial radioactive materials get into the atmosphere as a result of fission and fusion processes. More than 200 fission products are created with the fission of heavy nuclei, but the larger part of these quickly leave the atmosphere by sedimentation or precipitation, adhering to aerosols. The noble gases arising during fission represent an exception; they do not form compounds in the atmosphere and do not adhere to dust particles. Table 1 gives the noble gas isotopes with half lives longer than one hour arising in the fission of various heavy nuclei and their fission yields in percent (1 and 2). During fission there arise 16 isotopes of Kr and 20 isotopes of Xe; of these 10 are stable, the rest decay with short or very short half lives; the only exception is Kr-85, the half life of which is 10.76 years (3).

Kr-85 in the Atmosphere

The Kr-85 arising during nuclear weapon experiments gets into the atmosphere in its entirety; in the processing of reactor fuel elements a part of the active gas is captured for further use, but the rest gets into the atmosphere. Because of the long half life the Kr-85 can accumulate over the decades.

In 1955 S. Szalay and his colleagues (4) thought that the radioactive Kr isotopes arising during fission could be demonstrated by means of their radiation during production of Krypton gas. The enrichment of a radioactive material during manufacture could be experienced, but on the basis of half life measurements it could be identified as Rn-222. One could not demonstrate the presence of a fission product with the measurement method used.

In order to establish the atmospheric Kr-85 content the Krypton Factory in Miskolc provided us with samples of Kr gas. Kr gas is produced there by the Linde process (4) by enrichment (5) of the Kr content of air, having a volume part of $1.14 \cdot 10^{-6}$. On the basis of factory data the composition of the gas, by percent volume, is the following: 98.16% Kr; 1.18% Xe; 0.49% Ar; 0.16% N₂; 0.01% O₂; and 0.01% CO₂.

In order to establish how much of the Kr gas was Kr-85 we measured the beta radiation of Kr-85 ($E_{\max} = 0.67$ MeV). We used the internal gas counter method, by putting the radioactive gas directly into the GM tube. Knowing the effective volume of the counter tube it was possible for us to determine the specific activity.

In order to determine the effective volume we prepared GM tubes of three different lengths with the same final design; we filled the tubes with 90 Torr Krypton and 10 Torr isobutane gas and performed the measurement after complete decay of the Rn content of the gas. We used the factory Kr gas to fill the tubes without further purification. A detailed description of the design of the tubes and the determination of the effective volume can be found in a communique (6).

We performed the measurements on Kr gas from August 1954 and December 1966. We referred the measured activity to 1 cm³ of Kr gas in normal state (NTP) and to the time of collection, calculating with a T=10.76 years half life.

Figure 1 shows our measurement data, compared to the data of other authors (7-13). On the basis of the measurements of various authors one can establish a continuous increase in the Kr-85 activity; the activity doubled about every 3-4 years.

The Origin of Atmospheric Kr-85

The activity measured by us at the end of 1966 (22.4 decays/min cm³ Kr NTP) is about 20 times that of 1954. At present there are $1.8 \cdot 10^8$ Kr-85 atoms in 1 cm³ of normal state Kr gas, or 208 Kr-85 atoms in 1 cm³ of air.

Because of the long half life and the relatively slow increase in concentration it is just to presume that the Kr-85 is evenly distributed in the atmosphere. If we take the total mass of the atmosphere of the Earth as $5.3 \cdot 10^{21}$ g (14), then we get 121 kg Kr-85 as the Kr-85 content of the atmosphere, which corresponds to an activity of 47.2 MCi.

Knowing the fission yield of Kr-85 we can make an estimate of the quantity of fissioned material. The Kr-85 yield for fission processes according to the data in Table 1 varies between 0.127 and 0.87 percent depending on the fissioning material used and the energy of the neutrons producing fission. Since there has been no information about the ratio of various fissioning materials used it seemed appropriate to relate our measurements to fission of U-235 with thermal neutrons, thus we get the quantity of fissioning materials used in U-235 equivalents.

According to the most precise measurements at present the yield of Kr-85 in the case of fission of U-235 with thermal neutrons is 0.273% (15). The fissioning of material equivalent to 120 tons of U-235 would be necessary to create the 121 kg of Kr-85 determined by us, which represents a fission energy corresponding to 2,400 Mt TNT.

As of the end of 1962 nuclear weapons with an energy corresponding to about 510 Mt of TNT had been exploded, of which--according to estimates--the energy deriving from fission was equivalent to 193 Mt TNT and that from fusion was equivalent to 317 Mt (16). Thus, as of 1962, they used a total of fissioning material equivalent to 9.6 tons of U-235 in nuclear weapon experiments. These data have changed to practically a very small degree in the course of latter years because there have been only a few atmospheric nuclear weapons experiments by countries not party to the nuclear test ban agreement.

But according to our measurements the quantity of fissioned material is equivalent to at least 120 tons of U-235, thus more than 10 times the quantity arising from nuclear weapons experiments. These data are unambiguous proof that only a smaller part of the Kr-85 contamination of the atmosphere comes from nuclear weapons experiments and the larger part is from plants processing spent fuel elements. The same thing is proven by the fact that since the 1962 nuclear test ban agreement a further increase in activity can be established.

The total quantity of fissioned material established by us, equivalent to 120 tons of U-235, can be regarded as an under-estimate and the actual fissioned material can be regarded as more than this, because we did not take into consideration in our calculations that a part of the activity of the Kr-85 has already decayed, nor that a part of the Kr-85 created in reactors and during reprocessing certainly did not get into the atmosphere, nor that in many places the spent fuel elements are only stored for the time being and are not yet processed.

Although the Kr-85 activity in the atmosphere is increasing greatly and the present value ($11.5 \cdot 10^{-12}$ Ci/m³ air) will soon reach the value of average atmospheric Rn activity (10^{-10} Ci/m³ air), this does not represent a.

dangerous concentration radiobiologically. The maximum permissible concentration of Rn is $3 \cdot 10^{-9}$ Ci/m³ air, while in the case of Kr-85, $3 \cdot 10^{-7}$ Ci/m³ air is permitted (14).

From the viewpoint of industrial use the present contamination of Kr gas is not perilous, but it can no longer be used to fill ionization chambers, because its own radiation has increased to several times the cosmic background. Xenon can continue to be used for such purposes.

I would like to express my thanks to the Miskolc Krypton Factory (chief engineer Bela Balla, factory unit leader) for the cooperation offered in the course of the measurements.

1) Izotóp	2) Felezési idő	U-233	U-235	Pu-239	Th-232	U-238
		3) termikus neutronok	4) gyors neutronok			
Kr-83 ^m	1,86 h	0,48				
Kr-83	stab	1,17	0,544	0,29	1,99	0,4
Kr-84	stab	1,95	1,00	0,47	3,65	0,85
Kr-85 ^m	4,4 h		1,3		3,9	
Kr-85	10,76 a	0,58	0,273	0,127	0,87	0,153
Kr-86	stab	3,27	2,03	0,76	6,0	1,38
Kr-87	76 min		2,7 ^a			
Kr-88	2,8 h		3,7			
Xe-128	stab			$2 \cdot 10^{-1}$		
Xe-129	stab			$4 \cdot 10^{-4}$		
Xe-130	stab			$5 \cdot 10^{-3}$		
Xe-131 ^m	12 d		0,03			
Xe-131	stab	3,39	2,93	3,78	1,62	3,2
Xe-132	stab	4,64	4,38	5,28	2,87	4,7
Xe-133	5,27 d		6,62	6,91		
Xe-133 ^m	2,3 d		0,16			
Xe-134	stab	5,95	8,06	7,47	5,38	6,6
Xe-135	9,2 h	6,0	6,3	7,27		5,5
Xe-136	stab	6,63	6,46	6,63	5,65	5,9

Table 1. Noble gas isotopes with half lives longer than 1 hour arising in fission and their fission yields in percent (1 and 2).

KEY: (1) Isotope
 (2) Half life
 (3) Thermal neutrons
 (4) Fast neutrons

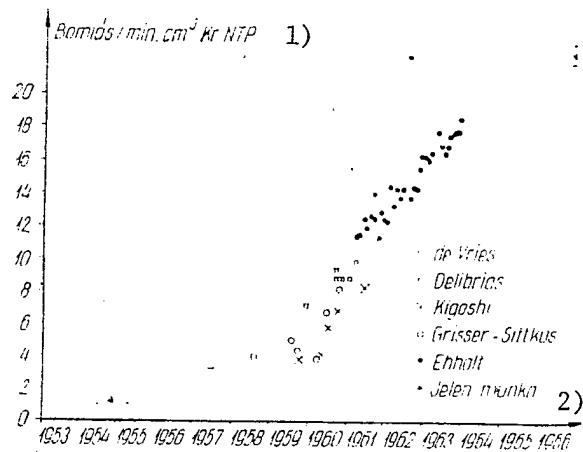


Figure 1. The increase in atmospheric Kr-85 activity in recent years.

KEY: (1) Decays per minute per cm³ Kr NTP
 (2) Present work.

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HUNGARY

MEASUREMENTS CONCERNING INCREASE OF ATMOSPHERIC RADIOKRYPTON CONTENT

Budapest IZOPTECHNIKA in Hungarian No 2, 1973 pp 96-103.

[Article by E. Csongor, Nuclear Research Institute of the Hungarian Academy of Sciences, Debrecen: "Measurements Concerning Increase of Atmospheric Radiokrypton Content"]

[Text] A paper read at a conference titled "Environmental Protection and Radioactive Standards" held in Sopron, 1-4 Nov 72, by the Radioanalytic Group of the Association of Hungarian Chemists and the Radioanalytic Work Committee of the Hungarian Academy of Sciences.

We can find many types of radioactive material in the atmosphere. Some of them are in the form of gas, others are suspended adhering to aerosols.

The radioactive material content of the atmosphere can be divided into two large groups:

1. Radioactive materials present independent of human activity:

a. gaseous products of natural radioactive materials; e.g., Rn, Tn and their decay products;

b. cosmogenic radioactive materials, which arise as a result of the mutual effect of cosmic radiation with the gases of the Earth's atmosphere (C-15, T, Ar-39, Be-7).

2. Radioactive contamination of the atmosphere deriving from human activity:

a. a large volume of artificial radioactive material, fission or activation products, arising as a result of nuclear weapon and other (primarily meteorological) experiments;

b. many, primarily gaseous, products due to the operation of reactors and primarily the processing and reprocessing of nuclear fuel elements.

If radioactive materials get into the atmosphere as a result of any of the processes mentioned above they quickly reach the surface of the Earth from

the troposphere by sedimentation or by precipitation, adhering to aerosols (with the exception of the noble gases, which stay in the atmosphere).

The fission of heavy nuclei produces 36 noble gas isotopes. Some of these are stable, some have short half lives, with the exception of Kr-85, which has a half life of 10.76 years.¹ Thus, alone among the fission products, this may appear as a lasting contaminant of the atmosphere.

Detecting and making a quantitative measurement of Kr-85 in the atmosphere represents a special analytical problem, partly because it is a noble gas and partly because the concentration of Kr gas itself in the atmosphere is very small (1.14 ppm).

Prior to the artificial production of fission it was not possible to detect the presence of Kr-85 in the natural background radiation experimentally. We can only estimate the quantity of Kr-85 created by natural means (for example by spontaneous fission or neutron acceptance by atmospheric Kr-85). One gets a value of $5 \cdot 10^{-6}$ pCi/m³ air as an upper limit.⁹ Thus Kr-85, as an atmospheric contaminant, was entirely negligible, with an atmospheric Rn concentration on the order of an average 100 pCi/m³ air, until the practical use of fission, thus as long as large amounts of Kr-85 did not get into the atmosphere as a result of nuclear weapons experiments and the nuclear industry.

Systematic measurements aimed at establishing the Kr-85 content of the atmosphere have been made since 1958. The first measurements were done by de Vries and Delibrias in France in 1954.^{2,3} Systematic studies to establish the Kr-85 content of the atmosphere have been conducted since 1958 in France^{4, 12}, since 1959 in Heidelberg (FRG)^{5,7,8,14}, since 1966 in two laboratories in the United States^{10,11,13} and also since 1966 in Debrecen^{15,16}.

Figure 1 contains a summary of the data published thus far. [See next page.]

A larger part of the data were obtained in internal gas filled proportional counters. The French group is using a 1.2 l volume counter, measuring the quantity of Kr gas introduced with a mass spectrometer. They add an Ar+CH₄ carrier gas to the Kr up to 740 torr and use 36 GM tubes as a protection counter.

The Heidelberg group uses an 11 cm³ proportional counter. The loading gas is 100-200 torr Kr, to which they add Ar+CH₄ to a total pressure of 700-720 torr. As a protection counter they use a hollow plastic scintillator.

The measurements in the United States are done with a scintillation counter. The Kr sample used for counting is taken directly from the air. They get several cm³ of Kr from several m³ of air, after multiple freezing, chemical sorption and baking procedures. They measure the activity either by putting a 20-30 mesh plastic scintillator scrap into a glass ampulla of several cm³ and putting the gas to be measured into this or by dissolving about 10 cm³

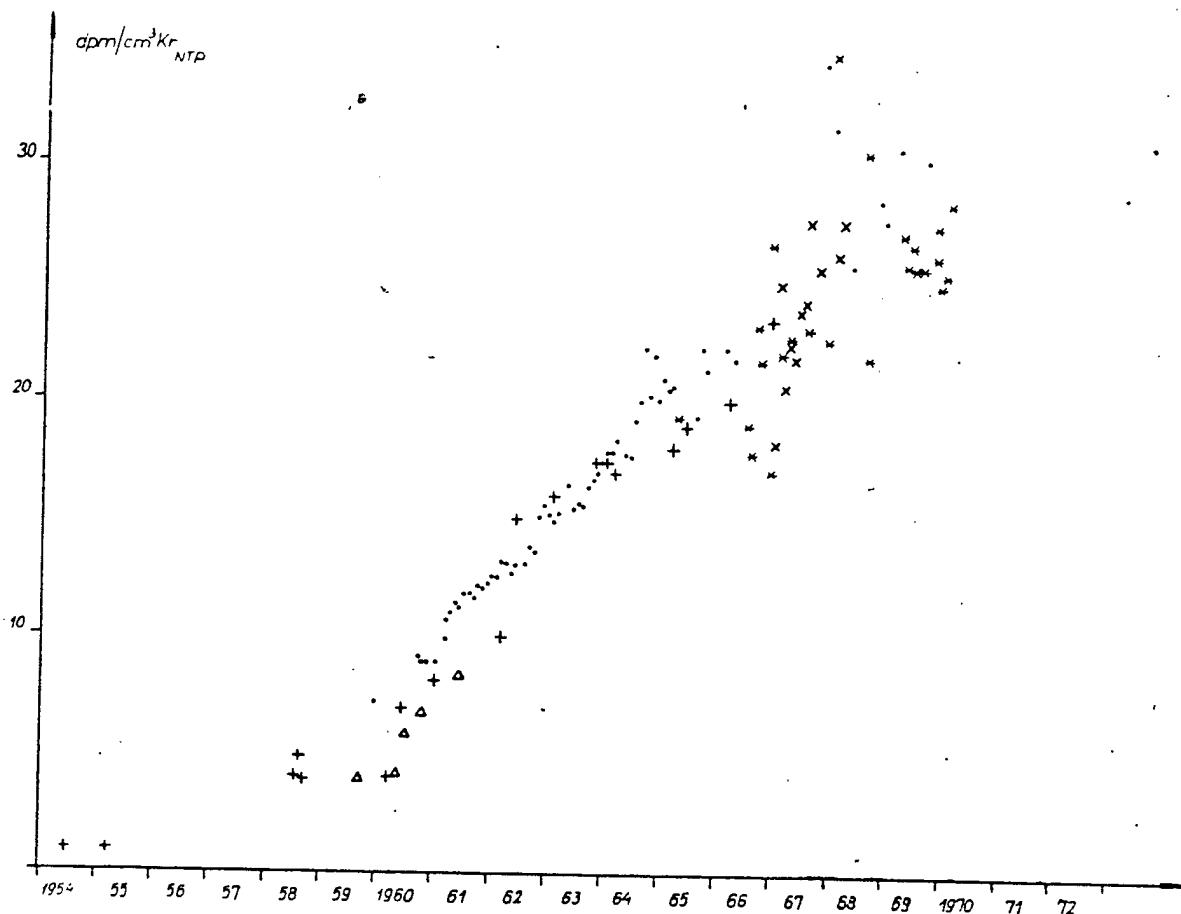


Figure 1. Atmospheric Kr-85 activity.

KEY: (1) (+) France
 (2) (.) FRG
 (3) (Δ) Japan
 (4) (*) United States

in about 25 cm³ scintillator liquid (the solubility of Kr in aromatic solvents is very good, 1 ml Kr/ml solvent).

The method I use to measure the atmospheric Kr-85 concentration is as follows.

I get the Kr gas samples from the Miskolc Krypton factory of United Incandescent, where the gas is produced by the Linde process by enriching the Kr content of the air, a volume part of $1.14 \cdot 10^{-6}$. In general the Kr content of the gas is around 98 percent, and we check its purity with a mass spectrometer. In the course of the manufacturing process the radon in the gas is enriched also ($T=3.8$ days) so all our measurements are done after the complete decay of the radon content of the gas, one month after the sample is taken.

We determine the Kr-85 content of the Kr gas by measuring its beta activity ($E_{\text{beta max}} = 0.67 \text{ MeV}$). We use a GM counter which we made ourselves for the measurement; figure 2 shows the design of the tubes. Since our goal is to measure the Kr-85 concentration we had to determine the effective volume of the tubes, so we prepared GM tubes of three different lengths (27.4 and 17.4 cm fiber length) with the same diameter (2.52 cm) and the same final design, and with these we determined experimentally the effective volume.

We fill the tubes with 90 torr Krypton and 10 torr isobutane gas; we use the factory Krypton gas to fill the tubes without further purification. We shield the GM tube with lead 68 mm thick. There is no need for a separate protection counter to decrease background radiation, because at a pressure of 90 torr the number of impulses per minute is about 10 times the background value.

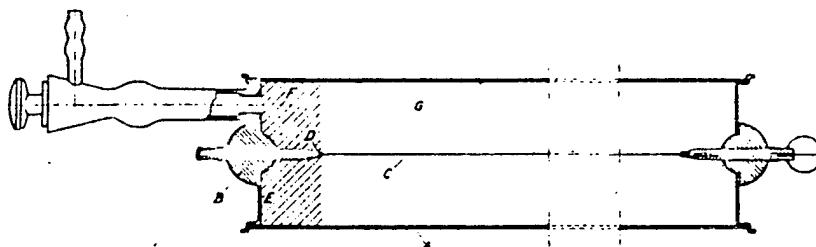


Figure 2. Cross section of GM tube.

KEY:

- (A) Electropolished copper.
- (B) Sealed glass bead.
- (C) 0.1 mm diameter W [tungsten] fiber.
- (D) Copper Capillary.
- (E) Copper sealing plate.
- (F) Insensitive end volume.
- (G) Sensitive volume.

Figure 3 [following page] shows the data for measurements done at ATOMKI [Nuclear Research Institute] together with the comparative data given earlier. It can be seen that they are in good harmony with the results obtained in other laboratories.

Since 1966 we have regularly requested samples from the Miskolc Krypton factory twice per year. The measured activity values are referred to 1 cm^3 NTP Kr gas and the time of collection of the sample.

One can establish from the measurement data a constant increase in atmospheric Kr-85. During the 5 years of the measurement the activity has increased by about 40 percent.

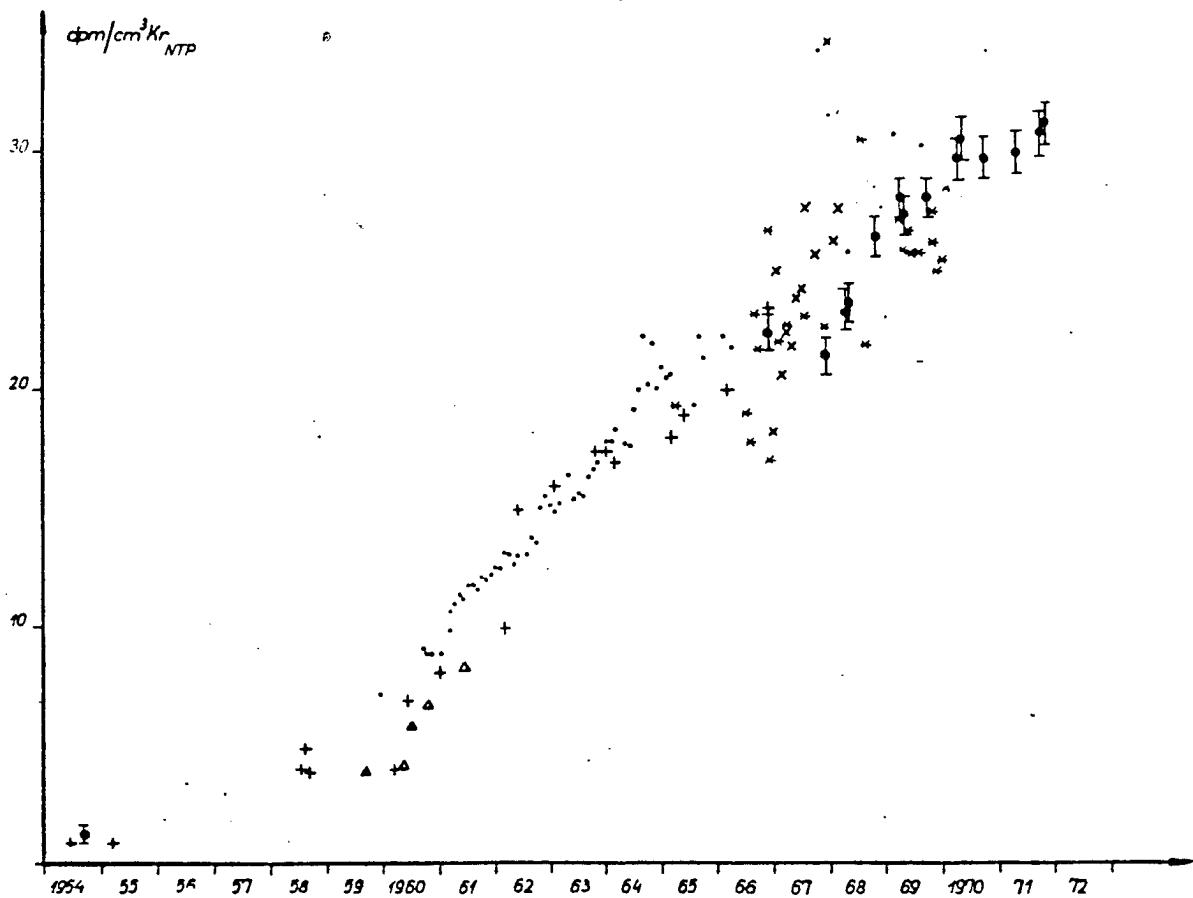


Figure 3. Increase in Kr-85 content of atmosphere. The figure shows the data for measurements done in Debrecen between 1966 and 1971 (o) compared to the foreign data published thus far (Figure 1).

Our activity values can be recalculated to other concentration units (Table 1), or, presuming that the Kr-85 is evenly distributed in the atmosphere, one can calculate the total number of Kr-85 atoms in the atmosphere and the total activity which can be attributed to them (at present 65.6 MCi). We can justly presume the even distribution of Kr because Kr is a noble gas and its half life is relatively long. Although the data were determined in various laboratories in the northern hemisphere and with various measurement methods the good agreement of the data given in Figure 3 can be regarded as experimental proof that Krypton is evenly distributed in the atmosphere. There are also measurement data¹² to the effect that the concentration of Kr-85 hardly changes with altitude to a pressure of 50 torr.

Table 1. Characteristic measurement data, in Debrecen, pertaining to the increase in atmospheric Kr-85 content.

Mérés ideje	dpm cm ³ Kr	pCi cm ³ Kr	pCi m ³ levegő	⁸⁵ Kr atomok száma a légkörben	⁸⁵ Kr tartalom MCi
1966.dec. 22,4 ± 0,7	10,1	11,5	0,86 . 10 ²⁷	47,2	
1968.okt. 26,4 ± 0,8	11,9	13,5	1,01 . 10 ²⁷	55,6	
1971.nov. 31,2 ± 0,9	14,2	16,2	1,2' . 10 ²⁷	65,6	
Évi növekedés			1		4

KEY:

- (1) Time of measurement
- (2) Air
- (3) Number of Kr-85 atoms in atmosphere.
- (4) Kr-85 content
- (5) Annual increase

Thus a constant increase in the Kr-85 concentration can be established from the measurement data. The next question is to clarify the origin of the Kr-85--What causes this increase in environmental contamination?

It was noted in the introduction that the concentration of Kr-85 appearing in the natural background is $5 \cdot 10^{-6}$ pCi/m³ air, or smaller than the present value by almost seven orders of magnitude.

If we compare the course of curve 1 with the curve of changes in time of fallout radioactivity coming from nuclear weapons experiments (Figure 4) we find two striking deviations (the measurement of fallout activity was done in our institute also, the results of the Debrecen measurements are shown in the figure):

- a. there is no correlation with the 1961-1962 series of nuclear weapons tests;
- b. when measuring Kr activity one does not observe the seasonal change characteristic of fallout activity (the spring peak).

These facts are qualitative proof that nuclear weapons tests are not the chief sources of atmospheric Kr-85.

We can get quantitative proof of this if we calculate how many Kr-85 atoms were put into the atmosphere by the fissioning material used in the nuclear weapons tests. It is known that in the course of the tests they used fissioning material equivalent to about 200 Mt TNT, which corresponds to

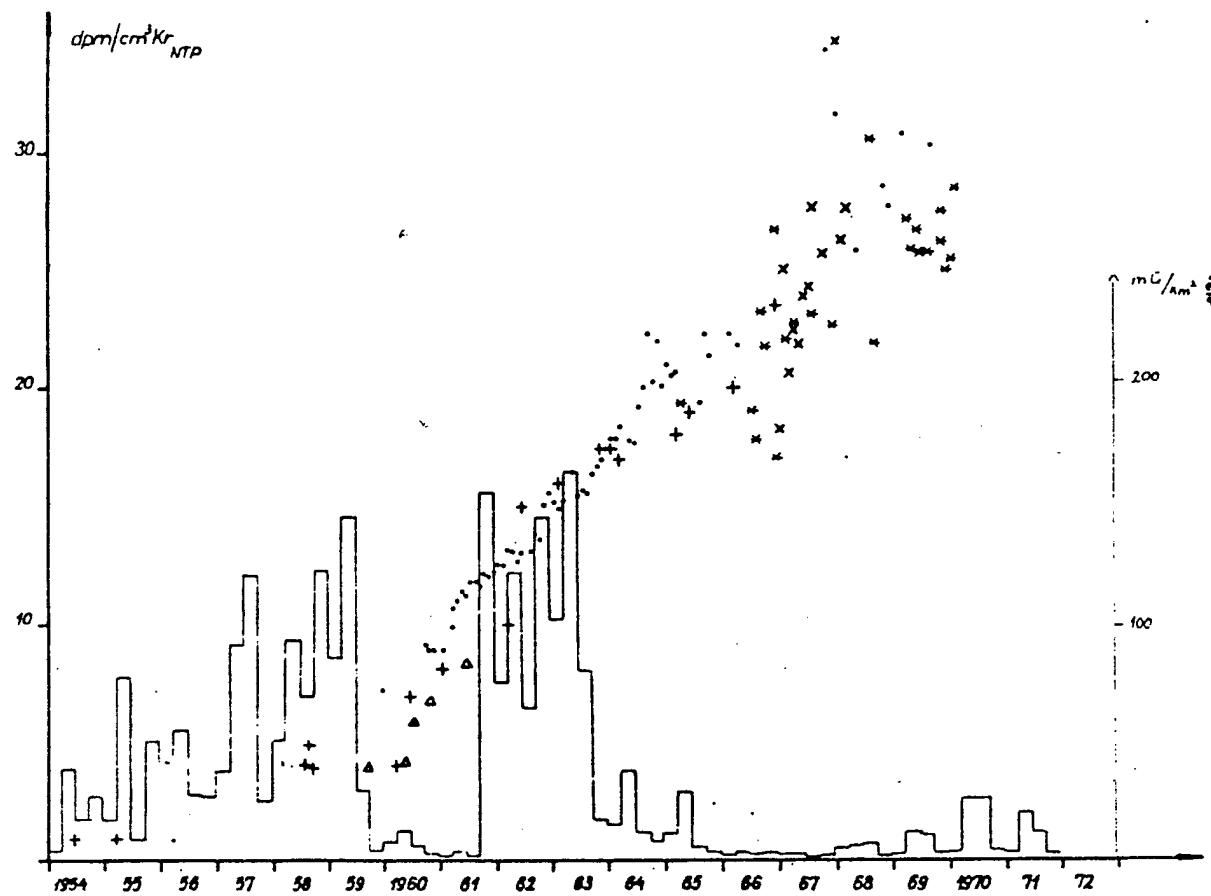


Figure 4. Increase in atmospheric Kr-85 activity, compared to changes over time in fallout radioactivity due to nuclear weapons tests. The figure gives the values for fallout activity measured at ATOMKI summing by quarter year in units of $\text{mCi}/\text{km}^2 \cdot \text{quarter year}$.

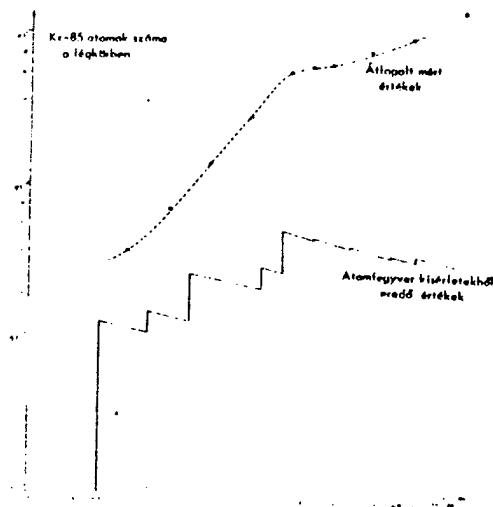


Figure 5. Lower curve: number of Kr-85 atoms coming from nuclear weapons tests (calculated with a half life of 10.7 years); upper curve: average activity values which can be fitted to the measurement times of Figure 3 expressed in number of atmospheric Kr-85 atoms.

roughly $70 \cdot 10^{24}$ Kr-85 atoms. Thus the concentration of atmospheric Kr-85 due to nuclear weapons tests was about 1 pCi/m^3 air at the beginning of the 1960's. In the lower curve of Figure 5 we can see the number of Kr-85 atoms deriving from nuclear weapons tests, the value of which was reduced practically by half in the past 10 years, since the large series of tests in 1961-1962. There have been only scattered French and Chinese nuclear weapons tests since the nuclear quiet agreement, and these resulted in a very tiny contamination increment.

The upper curve of the figure gives the average activity values, adjusted to the measurement points and expressed in the number of atmospheric Kr-85 atoms.

It can be seen from the figure that the present Kr-85 content of the atmosphere is more than 30 times what can be attributed to the nuclear weapons tests.

These data provide quantitative proof that the Kr-85 content of the atmosphere can be attributed almost exclusively to the nuclear industry.

Kr-85 is produced in reactors as a fission product, because of its long half life it decays only a little in reactor systems. The quantity of Kr-85 entering the atmosphere in the course of operating reactors does not exceed a value of $4 \cdot 10^3 \text{ Ci}$ per year; that is, an average concentration increase of 10^{-3} pCi/m^3 air per year due to operation. But since Kr is a noble gas it is difficult to segregate during reprocessing, and expensive with present technology. This is why the fuel element reprocessing sites now release Kr-85 into the atmosphere, not rarely releasing 10^3 - 10^4 Ci Kr-85 into the atmosphere on some days, as a result of which the annual increase will be about 1 pCi/m^3 air, or 4 MCi/year (Table 1).

Two observations can be made in connection with the release of Kr-85:

1. Since release at the reprocessing plants is not continuous this is the chief reason for the spread in the measurement data, which appears especially in the measurement data in the United States and more recently in the West German data.
2. The other observation in connection with the release of the gas is of interest from the viewpoint of environmental contamination and radiation protection. Kr-85, as a noble gas, spreads throughout the atmosphere without mutual effects and causes radioactive contamination not only locally, in regions with a developed nuclear industry, but throughout the entire atmosphere without mutual effects and causes radioactive contamination not only locally, in regions with a developed nuclear industry, but throughout the entire atmosphere of the Earth. The present concentration of Kr-85 represents

a surface beta dose of somewhat more than 0.03 mrem/year; thus, at present, it does not represent a substantial radiation burden as compared to the environmental dose of 100-150 mrem/year. But the measurement data indicate a swift increase in Kr-85 contamination. Taking into consideration the expected development of the nuclear industry and the increasing use of fissioning materials it has been estimated¹⁷ that in 2060 the concentration of Kr-85 in the atmosphere will be about 50,000 pCi/m³ air, which corresponds to a dose of 50-100 mrem/year (Figure 6), thus the dose burden from Kr-85 will double the present environmental burden.

From this single datum it can be seen how desirable and necessary it is to develop a segregation process with which the Kr-85 can be extracted in plants processing reactor fuel elements so that it can be used for a useful purpose instead of contaminating the atmosphere.

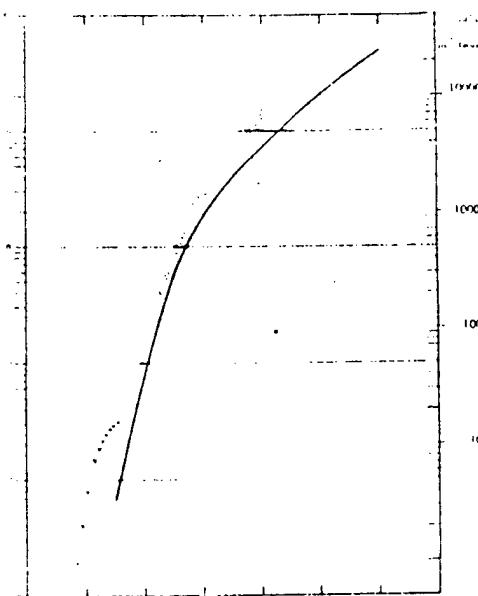


Figure 6. Estimate by Coleman and Liberace¹⁷ of the increase in atmospheric Kr-85 content between the years 1970 and 2060 and of the annual dose burden deriving from this. The figure shows the average value of western European (x) and Debrecen (.) measurement data.

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HUNGARY

MEASUREMENTS OF ATMOSPHERIC KR-85 CONCENTRATION AT ATOMKI FROM 1966 to 1977

Budapest ATOMKI KOZLEMENYEK in Hungarian No 21, 1979, pp 1-12

[Article by Eva Csóngor, ATOMKI Debrecen: "Methods of Measuring Atmospheric Kr-85 Concentration and the Measurements Done at ATOMKI Between 1966 and 1977"]

[Text] The author first provides a review of the various methods used thus far to measure the Kr-86 concentration and of enrichment and measurement procedures. In the second part she reports on the measurements done regularly at ATOMKI since 1966. The high purity Kr gas samples are obtained from the Krypton Factory in Miskolc. The measurements are done with an internal gas filled GM counter tube with an effective volume of 136 cm³ which filled with 90 torr Kr gas and 10 torr isobutane [the English summary adds: as quenching gas]. The communiqué contains a summary of data for measurements done since 1966. After an initial quick increase the Kr-85 concentration assumed almost steady values in the 1970's; the present concentration level is about 18 pCi/m³ air, which corresponds to an accumulated value of 70MCi in the entire atmosphere. On the basis of the stabilized value it was calculated that the reprocessing plants now operating release Kr-85 gas with an activity of 5 MCi into the atmosphere each year.

Kr-85 is a radioactive fission product which decays with beta emission with a half life of 10.7 years. Its presence in the atmosphere has been demonstrated beginning in the 1960's, when it entered the atmosphere in significant quantities in the course of atmospheric nuclear weapons tests, the operation of reactors and primarily the chemical processing (reprocessing) of burned out fuel elements (Figure 1). Compared to the above K4-85 sources the dose represented by Kr-85 appearing in the natural background radiation represents a tiny dose; Kr-85 is produced by the spontaneous fission of natural Uranium and as a result of activation of the stable Kr isotopes of the atmosphere by neutrons of cosmic origin.

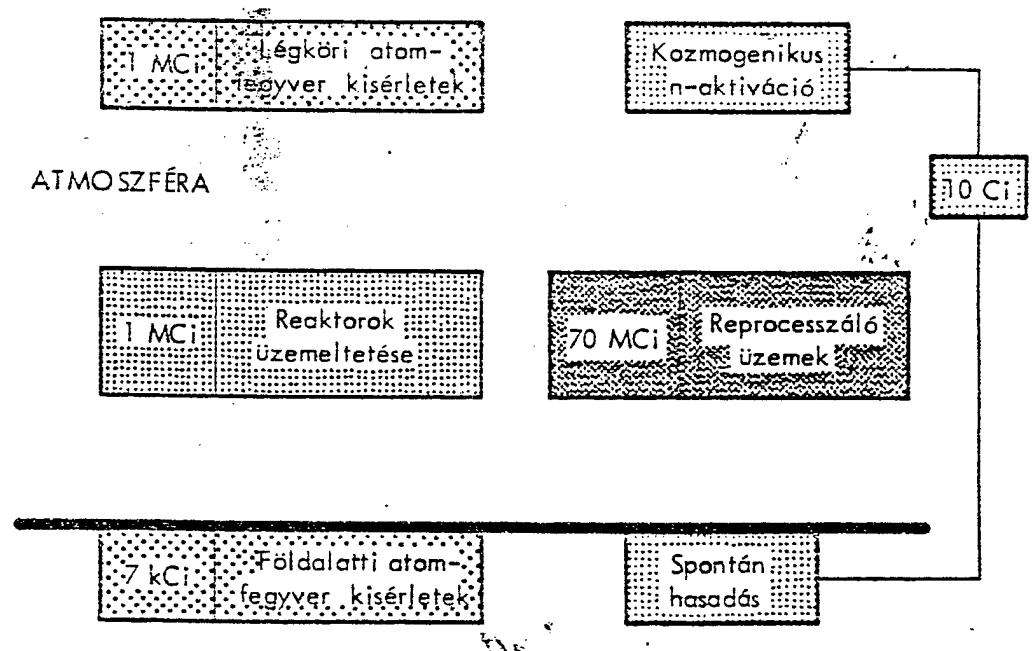


Figure 1. The sources of atmospheric Kr-86 and the activities accumulated up to 1977.

- 1 MCi--atmospheric nuclear weapons tests
- 1 MCi--operation of reactors
- 70 MCi--reprocessing plants
- 10 Ci --cosmogenic n-activation and spontaneous fission (combined)
- 7 kCi--underground nuclear weapons tests

The most significant of the sources mentioned above are the reprocessing plants which at present release into the atmosphere in their entirety the gas phase fission products (Kr, Xe, I, T, C-14). Of these Kr-85 has the greatest significance from the viewpoint of the radioactive contamination of the atmosphere for two reasons:

- a. Since Kr is a noble gas and its solubility in water is extraordinarily low ($1.8 \cdot 10^{-6}$ g Kr/g water) it remains practically entirely in the atmosphere; it moves together with the air masses both horizontally and vertically, mixing quickly, and can be found in an even distribution in the entire atmosphere.
- b. Because of its relatively long half life it accumulates to a large degree in a few decades in the atmosphere and because of the planned development of the nuclear industry one must reckon with a further increase in the concentration.

The present concentration of Kr-85 in the atmosphere is 0.67 Bq/m^3 air (18 pCi/m^3 air), which is equivalent to an accumulated activity of $2.6 \cdot 10^{18} \text{ Bq}$ ($\sim 70 \text{ MCi}$).

Regular measurement of the Kr-85 concentration is justified partly because one can conclude from the atmospheric Kr-85 concentration the quantity of fissioning material reprocessed and partly because Kr-85, as a radioactive environmental contaminant, represents a radiation burden for the populace.

Methods of Measuring the Activity of Atmospheric Kr-85

Kr-85 decays with beta emission; the maximum energy of the beta particles is 0.68 MeV. The beta radiation is accompanied, in a ratio of 0.43 percent, by gamma radiation with an energy of 0.514 MeV.

Measurement of its activity takes place most frequently with detection of the beta rays; measurement with gamma radiation can be used only at very high activity levels because of the low branching ratio.

Table 1. Kr-85 concentration levels occurring in the atmosphere.

	Bq/m ³ levegő ⁽¹⁾	pCi/m ³ levegő
Jelenlegi átlag (1977) ⁽²⁾	0,67	18
Maximálisan megenged- ⁽³⁾ hető koncentráció (MPC)	1,1.10 ⁴	3.10 ⁵
Reprocesszáló üzem ki- ömlő gáza [1] ⁽⁴⁾	3,7.10 ⁷	10 ⁹

KEY: (1) air
 (2) Present average (1977)
 (3) Maximum permissible concentration
 (4) Gas emitted by reprocessing plants (1)

Table I provides information about the Kr-85 concentration values occurring in the atmosphere.

1. Direct Measurement of the Activity of Atmospheric Samples

In the event of sufficiently great activity the Kr-85 activity of the air sample can be measured directly with an ionization chamber, and window counter tube, internal gas filled GM tube or proportional counter. The minimal detectable concentration for the above devices is $7.2 \cdot 10^3$ - $1.8 \cdot 10^3$ Bq/m³ ($2 \cdot 10^5$ - $5 \cdot 10^4$ pCi/m³) (2); thus this measurement technique can be used to measure concentrations only in the immediate vicinity of the reprocessing plant.

2. Methods of Taking Kr Samples

The activity level of Kr-85 in the atmospheric air is 0.67 Bq/m^3 (18 pCi/m^3 , that is 39 decays/min m^3 air), which is lower by about four orders of magnitude than can be indicated by direct measurement. Thus, for measurement purposes, the Kr must be concentrated by at least this amount.

Kr gas is present in normal atmosphere in a volumetric concentration of 1.14 ppm, $14 \text{ cm}^3 \text{ Kr/m}^3$ air; that is, complete extraction of the Kr gas means an enrichment of 10^6 times.

Concentration represents a problem partly because, for the above reasons, one must start with a relatively large quantity of air and partly because Kr is a noble gas and thus enrichment of it is not possible with the customary chemical methods; the Kr must be separated from the air with physical methods through adsorption on active carbon or a molecular filter (MS); with gas chromatography, low temperature distillation, etc.

Since the 1940's the extraction of Kr from air on an industrial scale has been a solved problem.

The first measurements of the Kr-85 content of the atmosphere were published from laboratories, and regular measurements were started, where the concentrated gas samples were provided by air liquefaction plants or plants producing high purity Kr gas. Thus measurements began in France by Delibrias et al (3), in the FRG by Griesser and Sittkus (4), by Ehhalt et al (5) and by Schroder et al (6), in Hungary by Csengor (7), in the USA by Shuping et al (9) and in the Soviet Union by Tertysnik et al (9).

With the spread of cryogenic technology there was a proliferation of measurement sites where air samples were taken and the measurement of Kr-85 takes place at the same sites.

In the course of developing enrichment techniques they used some combination of the physical methods mentioned earlier to separate the Kr and in the final phase the remnant non-noble gases were separated from the Kr sample chemically (Pannetier, 10, Sax et al, 11, and Cummings et al, 12).

In what follows we will describe a few separation methods of historic, theoretical or practical interest.

Pannetier (1) started with a 5 m^3 air sample and used adsorption on 5A type molecular filters (MS) and desorption with gradual warming to extract the Kr. He used a 500 degree C Zr-Ti furnace for final chemical purification of the Kr gas. Then the Kr was collected on active carbon and taken from here to a proportional counter. He could extract 30-80 percent of the entire Kr content of the sample and this yield was determined for every sample with a mass spectrometer.

Sax et al (11) collected three m^3 of air in a plastic sack in the field. In the laboratory the gas was adsorbed on active carbon and a 5A molecular

filter and only the fraction containing the Kr was retained with fractionated desorption. This was freed of remnant O_2 with a Cu furnace and of N_2 with liquid Li. He finally got 2-3 cm^3 of Kr gas with a yield of 90 percent. The yield was determined with a mass spectrometer.

Cummings et al (12) also used active carbon and a molecular filter for separation of the Kr and separated the gas fractions by means of chromatography. The final N_2 traces were removed with a Ti furnace. They used the short half life Kr-83 m tracer as an internal standard to determine the yield. This method gave a yield of 80-90 percent.

Stevenson and Johns (13) were the first to work out a process which produces high purity Kr under laboratory conditions without chemical purification. A 1 m^3 air sample is taken in the field; the gas is first absorbed on cooled active carbon in a vacuum system and the largest part of the O_2 and N_2 is driven out with He gas. The Kr with remnant gases is put in a 5A MS chromatographic column at the temperature of liquid N_2 . By increasing the temperature the gases are selectively desorbed from the column. During desorption the fraction containing the Kr is followed with a heat conductive cell. The process is repeated two times in the interest of better separation. In the final phase they measure the volume and pressure of the Kr and the yield is determined volumetrically, knowing the volume of the initial air. Kr yields of 70-80 percent can be obtained with their method.

More recently Johns (14)--on the basis of the principle described above--has developed portable equipment which can be used in the field. This is suitable for swift separation and measurement of atmospheric Kr, Xe and CH_4 . The separation equipment consists of several active carbon adsorption and chromatographic (5A MS) columns. In 2.5 hours it separates the Kr, Xe and CH_4 gases from a 1 m^3 air sample with yields of 85, 70 and 40 percent respectively. The activity of these is measured in the field with a liquid scintillation counter (point 1.B.3). The minimal detectable concentration with his equipment is 0.08 Bq/m^3 (2 pCi/m^3).

Equipment operating on a principle similar to the separation technique of Stevenson and Johns (13) is used in a number of laboratories also.

AIRCO Inc has developed equipment suitable for taking two samples per day for meteorological experiments (15). They achieve an enrichment of 10^4 times at the place of taking the sample by means of continual liquefaction of the incoming air using liquid N_2 (25 l/day), letting the more volatile components of the air boil away. They collect 5-10 cm^3 of Kr in 900 cm^3 samples from which they produce Kr with a purity of 99.98 percent in a central laboratory by means of gas chromatographic separation. Activity measurement is with a GM tube.

To sum up the sampling methods:

--either measurement is done on high purity samples produced industrially, --or they process 1-5 m^3 air samples under laboratory conditions with yields of 50-90 percent; they get 0.5-5 cm^3 of gas as a final product, which is measured with various types of detectors.

3. Measuring the Activity of Enriched Kr Samples

a. The first atmospheric Kr-85 concentration measurements were done with internal gas filled proportional or GM counters.

Delibras et al (3), Griesser et al (4), Csongor (7), Ferber et al (15) and Kigoshi (26) used high purity Kr gas directly as the filling gas.

Ehhalt et al (5), Pannetier (10), Schroder et al (6) and Stockburger et al (16) used a CH_4 carrier gas with the Kr in a proportional counter. The Kr concentration in the sample is reduced because of the carrier gas. Measuring such a low activity level can be done practically only if an anticoincidence protection is used for suitable reduction of the background.

Measurement with an internal gas filled counter requires high gas purity and special measuring equipment but at present the greatest sensitivity can be obtained with these detectors; the minimal detectable concentration in air is $0.02 \text{ Bq}/\text{m}^3$ ($0.5 \text{ pCi}/\text{m}^3$).

b. Today they most commonly use liquid scintillation counters for swift, routine determinations. Their advantage is that special equipment is not needed for the measurement, as in the case of internal gas filled counters, but rather all the equipment can be purchased as factory series instruments.

The spread of the liquid scintillation measurement method was made possible by the fact that the solubility of Kr in scintillation liquids based on toluol is extraordinarily good ($\sim 1 \text{ ml Kr}/\text{ml solution}$).

Shupine et al (8) were the first to succeed in getting a sufficient quantity of Kr gas into the scintillation liquid. They first put the Kr gas into the scintillation measuring vessel, measured the pressure of the gas, then filled the measuring vessel with the scintillation liquid from an injection syringe. They achieved a counting efficiency of 92 percent. The minimal detectable concentration with this method is $0.04-0.08 \text{ Bq}/\text{m}^3$ ($1-2 \text{ pCi}/\text{m}^3$).

Another measurement method has been developed in the area of scintillation measurement technology also. For example, Sax et al (11) put the Kr gas into a measurement vessel filled with a fine granular plastic scintillator; the efficiency was 94 percent and the minimal detectable concentration was $0.04 \text{ Bq}/\text{m}^3$ ($1 \text{ pCi}/\text{m}^3$).

Use of the measurement method developed by Shuping et al (8) is the most extensive in practice.

To sum up the Kr-85 activity measurement methods; in general two detector types are used most frequently:

--either internal gas filled proportional or GM counters, which are special devices. With these the minimal detectable concentration is $0.02 \text{ Bq}/\text{m}^3$ ($0.5 \text{ pCi}/\text{m}^3$).

--or liquid scintillation counters, which are suitable for fast, routine measurement of large numbers of samples. Their efficiency is 92 percent and their sensitivity is $0.04\text{--}0.08 \text{ Bq/m}^3$ ($1\text{--}2 \text{ pCi/m}^3$).

C. Measurement of Atmospheric Kr-85 concentration at the ATOMKI Between 1966 and 1977

1. Measurement Method and Results

The initiation of measurements in Hungary was made possible by the fact that we could get high purity Kr gas samples from the Miskolc Krypton Factory of the United Incandescent Lamp and Electric Company, where the gas is produced by the Linde process through enrichment of the Kr content of air, having a part volume of $1.14 \cdot 10^{-6}$. The purity of the gas is 99 percent; its purity is regularly determined with a quadrupole mass spectrometer. They also use the mass spectrum to check that isotope fractionization does not occur in the course of the enrichment process, that the original stable isotope ratios remain (23 and 27). Radon (a half life of 3.8 days) is also enriched in the gas during the manufacturing process, so we do the measurements after the complete decay of the radon content of the gas, one month after the sample is taken.

Two samples each are made available to us regularly, in the spring and fall of every year on a regular basis since 1966, by the Krypton Factory, where they put gas into one bottle daily for a month (a monthly average) and put into another bottle a sample from one week's collection. The efficiency of the enrichment process is 99 percent.

We determine the Kr-85 content of the Kr gas by measuring its beta activity. We fill a GM counter tube of our own design, with an effective volume of 136 cm^3 (7), with 90 torr Kr and 10 torr isobutane gas; we use the factory Kr gas to fill the counter tube without further purification. We shield the counter tube with 58 mm of lead. The activity level measured (500 cpm) is about 10 times the background, so there is no need for anticoincidence protection to decrease the background. The measurement time for the sample is 10 hours.

We relate the measured activity values to 1 cm^3 normal state Kr gas and the time the sample was collected. The precision of the measurements is 2 percent, deriving from the statistical error of counting and error in determining the effective volume. The minimal detectable concentration with the equipment is 0.02 Bq/m^3 (0.5 pCi/m^3 air), which coincides with the sensitivity level of the most sensitive equipment used today.

Table II and Figure 2 contain the measurement data.

2. Conclusions

On the basis of the measurement data published in Table II it can be established that the atmospheric Kr-85 concentration increased between 1966 and

Table II. Kr-85 concentration in the atmosphere between 1966 and 1977 measured in Debrecen (7 and 19-23).

Minta gyűjtésének ideje (1)	(2) Koncentráció Bq/m ³ pCi/m ³ (3) hiba \pm 2 %	Minta gyűjtésének ideje (1)	(2) Koncentráció Bq/m ³ pCi/m ³ (3) hiba \pm 2 %
1966.dec.6.-dec.7.	0,43 11,5	1973.nov.10.-dec.7.	0,61 16,4
1967.nov.17.-nov.20.	0,41 11,1	dec.2.-dec.7.	0,59 15,9
1968.ápr.20.-máj.13. máj.11.-máj.13.	0,45 12,1 0,45 12,2	1974.máj.4.-máj.30. máj.22.-máj.30.	0,56 15,0 0,53 14,3
okt.5.-nov.1.	0,51 13,7	nov.20.-dec.11.	0,63 17,0
1969.ápr.20.-máj.16. máj.10.-máj.16.	0,54 14,5 0,52 14,1	dec.6.-dec.11.	0,58 15,6
okt.4.-okt.26.	0,54 14,5	1975.ápr.22.-máj.14. máj.7.-máj.14.	0,63 17,0 0,59 15,8
1970.márc.13.-ápr.9. ápr.1.-ápr.9.	0,57 15,4 0,59 15,8	nov.23.-dec.3.	0,60 16,1
szept.23.-okt.15.	0,57 15,4	1976.ápr.27.-máj.19. máj.14.-máj.19.	0,62 16,7 0,63 16,9
1971.márc.31.-ápr.22. okt.31.-nov.25.	0,57 15,5 0,59 15,9	nov.7.-dec.3.	0,68 18,2
nov.15.-nov.25.	0,60 16,2	nov.26.-dec.3.	0,62 16,7
1972.ápr.21.-máj.12. nov.16.-dec.8.	0,57 15,4 0,60 16,1	1977.márc.17.-ápr.5. márc.31.-ápr.5.	0,61 16,5 0,62 16,7
1973.ápr.29.-máj.24. máj.20.-máj.24.	0,58 15,6 0,57 15,3	nov.15.-dec.9. dec.1.-dec.9.	0,64 17,3 0,65 17,6

KEY: (1) Time of collecting sample
 (2) Concentration
 (3) Error

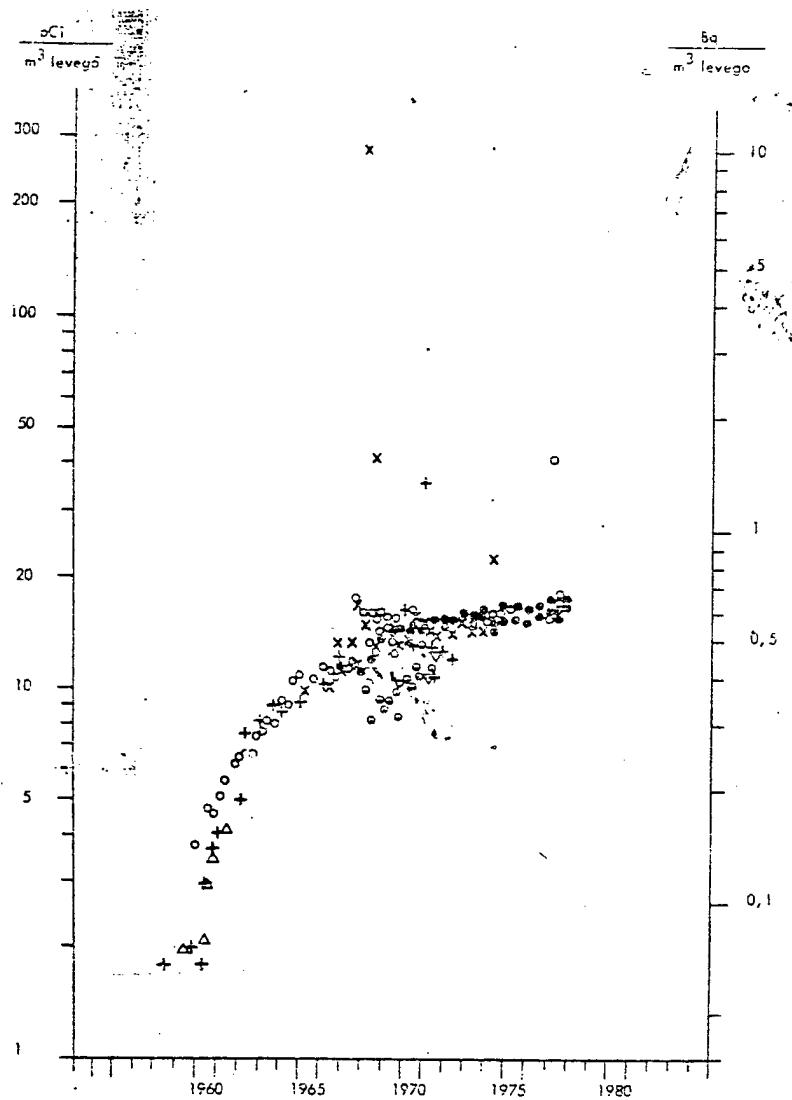


Figure 2. Measured Kr-85 concentrations [3]-[11], [15], [16], [18]-[26].
 NORTHERN HEMISPHERE: Germany: \circ ; France: $\nabla +$; Debrecen: \square ;
 USA: \times ; Japan: Δ ; SOUTHERN HEMISPHERE: \bullet .

1970 with an average annual value of 0.04 Bq/m^3 (1 pCi/m^3), then practically stabilized after 1971, and again increased very slightly from 1974.

Figure 2 also shows the measurement data of other authors published thus far (3-11, 15, 16, 18 and 24-26).

The following conclusions can be drawn from the combined measurement data:

--The Kr-85 concentration values show the same levels and trends in the entire northern hemisphere--after the initial fast increase the increase has slowed beginning in the 1970's. The reason for this is that the number of reprocessing plants has not increased; indeed, at present not one of the industrial reprocessing plants is in operation in the United States (17) and the burned out fuel elements are being stored without reprocessing. The explanation for this can probably be sought in the fact that they have stopped obtaining Pu (serving primarily military purposes) from the fuel elements. In western Europe, however, several smaller capacity reprocessing plants are in operation (17).

--The almost constant concentration value also means that the annual quantity of Kr-85 released by the reprocessing plants now operating hardly exceeds the value by which the Kr-85 content of the atmosphere decreases each year by virtue of radioactive decay. In the event of an accumulated activity of about $2.6 \cdot 10^{18} \text{ Bq}$ (70 MCi) that is $0.18 \cdot 10^{18} \text{ Bq/year}$ (5MCi/year). From these data one can calculate that the quantity of fuel elements reprocessed per year in recent years is about 1,000 tons (5 kCi Kr-85/t fuel elements [17]).

--The large spread appearing in some data for measurements done in the northern hemisphere (16, 11, 15 and 24) is caused by the fact that the reprocessing plants do not release Kr-85 into the atmosphere continually, thus very high values in the Kr-85 concentration can be measured from time to time in the immediate vicinity of the reprocessing plants. The released Kr moves together with the air masses generally from west to east, circling the Earth for the first time in about 30 days and requiring about 2 years for north-south mixing. Thus the concentration of Kr-85 in the first 2 years depends somewhat on the latitude; thereafter the Kr-85 can be found distributed evenly in the entire atmosphere. This also explains why samples from the southern hemisphere (24) show a somewhat lower activity level.

--The measurement data in Figure 2 can be regarded as data for a meteorological experiment done with a Kr-85 tracer, on the basis of which it can be established that Kr-85 is distributed evenly around the world in the atmosphere and thus it represents a constantly increasing radioactive contamination for the entire world.

Model calculations done on the basis of the measurement data give a one year half time as a measure of even distribution in the northern hemisphere, where the reprocessing plants operate, but with a delay of about 2 years the Kr-85 appears in the southern hemisphere also as a contaminant with the same concentration.

Thus Kr-85 is present in the atmosphere as a radioactive environmental contaminant which, due to its beta radiation, represents a skin surface dose burden of 0.03 mrem/year per person. This present value of the dose is insignificant in comparison with the doses received due to the natural background and medical treatment.

As a final conclusion we can establish that measurement of the concentration of atmospheric Kr-85 can be done with goals of two sorts.

1. We want to determine the Kr-85 concentration values corresponding to an even distribution as a function of the calendar year (see Figure 2).. This is the minimal concentration possible, a concentration smaller than which cannot occur in the atmosphere today (the radiation burden aspect).

In order to determine the yearly average it is enough to measure a few samples per year, samples which have been collected over a longer time. But this method of measurement can be used only where there are no reprocessing plants nearby.

The method used in Hungary corresponds to this goal; it is suitable for observing average activity levels, due to its sensitivity and precision.

2. If, however, we want to follow fluctuations in the Kr-85 concentration in the immediate or more distant vicinity of a reprocessing plant or nuclear power plant (the nuclear industry or meteorological aspect) then we must have local sampling and, if possible, local enriching equipment so that samples can be taken at least every 10 days. Measuring the separated gas samples can be done at the site where the sample is taken or in a measurement center.

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Professional reader: Sandor Bohatka

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